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Masahiro Yamada ^a , Hideharu Ueki ^a , Satoshi Makino ^a , Hirohumi Fukai ^b & Yasushiro Nishioka ^{a b} ^a College of Science and Technology, Nihon University, Funabashi-shi, Chiba, Japan ^b Research Center for Micro-Functional Devices, Nihon University, Funabashi-shi, Chiba, Japan

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Influence of Ambient Pressure on the Performance of Pentacene Organic Thin Film Transistors with Polyimide and SiO₂ Gate Insulators

Masahiro Yamada¹, Hideharu Ueki¹, Satoshi Makino¹, Hirohumi Fukai², and Yasushiro Nishioka^{1,2}

¹College of Science and Technology, Nihon University, Funabashi-shi, Chiba, Japan

²Research Center for Micro-Functional Devices, Nihon University, Funabashi-shi, Chiba, Japan

The reliability of Organic Thin Film Transistors (OTFTs) is of special interest. Here, the instability of the pentacene OTFTs with different gate insulators of SiO_2 and polyimide on the measurement ambient pressures has been studied. The gate insulators are thermally grown SiO_2 or spin coated polyimide films on heavily doped n^+ silicon wafers. The semiconductor layer of pentacene and the Au layer for the source and drain electrodes were deposited by thermal vacuum deposition. P-channel OTFTs with $Si/SiO_2(300\,\mathrm{nm})/\mathrm{pentacene}(70\,\mathrm{nm})/\mathrm{Au}$ and $Si/\mathrm{polyimide}(320\,\mathrm{nm}-620\,\mathrm{nm})/\mathrm{pentacene}(70\,\mathrm{nm})/\mathrm{Au}$ structures have been fabricated and the transistor electrical characteristics in the air and in the vacuum pressure below $1.3\,\mathrm{Pa}$ were measured. As a result, $I_D\text{-V}_{DS}$ characteristics of the transistor with the gate insulator of SiO_2 exhibited notable changes depending on the pressure. On the other hand, the transistor with the polyimide gate insulator exhibited the minimal influence from the ambient pressure. These differences in the ambient pressure dependence are attributed to the difference in the gate insulator/pentacene interfaces.

Keywords: gate insulator; organic thin film transistor; pentacene; polyimide

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Address correspondence to Yasushiro Nishioka, Department of Precision Machinery Engineering, College of Science and Technology, Nihon University, 7-24-1 Narashinodai, Funabashi-shi, Chiba 274-8501, Japan. E-mail: nishioka@eme.cst.nihon-u.ac.jp

INTRODUCTION

Organic Thin Film Field Effect Transistors (OTFTs) have been actively studied, and those are expected to be used for flexible displays, liquid crystal and other electric devices [1–3]. Recently the performance of OTFT has been improved considerably, and the field effect mobility of an organic single-crystal transistor is now reaching $40\,\mathrm{cm^2/Vs}$ [4]. However, there should be many issues to use those OTFTs in products. For example, reliability of those devices is one of the key issues for the practical uses of OTFTs. Recently, Yokoyama et al. have reported that an n-channel OTFT with the Si/SiO₂/ perfluoropentacene/Au structure shows the long period and irreversible degradation of the carrier mobility when the devices were stored in dry O₂ [5]. They attributed to the electron mobility degradation behaviors of the OTFTs to the oxidation of the perfluopentacene when the OTFTs were stored in the air [5]. On the other hand, Matsushita *et al*. reported that an n-channel OTFT with the Si/SiO₂/F₁₆CuPC/Au structure exhibits nearly reversible changes of the electron carrier mobility when the OTFTs were measured in the air and in the vacuum alternately [6]. They speculated that some chemical species in the air would penetrate into the SiO₂/F₁₆CuPC interface when the devices were stored in the air, and that those chemical species diffuse out from the interface in the vacuum.

On the other hand, OTFTs used in flexible displays and other flexible devices need to be fabricated on flexible gate insulators and substrates, and many reports on OTFTs with organic gate insulators such as polyimide can be found everywhere [7]. In addition, pentacene thin films have been most actively studied because of its very high hole mobility. However, reports on the reliability of p-channel OTFTs with polyimide gate insulator are not very many to the best of our knowledge. Therefore, we fabricated p-channel OTFTs with the Si/polyimide/pentacene/Au and Si/SiO₂/pentacene/Au structures, and the transistor characteristics in the air and in the vacuum are compared.

EXPERIMENTAL

Figure 1 shows the schematic cross-sectional view of the pentacene OTFT investigated in this study. Here, two kinds of gate insulators, SiO_2 and polyimide (PI), were prepared. The 300 nm-thick SiO_2 was thermally grown on heavily doped n^+ silicon wafers with the resistivity less than 0.02 ohm cm. The PI was the CT4112 provided by Kyocera Chemical Company. The liquid precursor of the PI was dropped on the

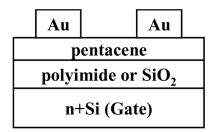


FIGURE 1 The schematic cross-section of the pentacene OTFT.

heavily doped n⁺ Si substrate, and was spin-coated with rotation speeds of 800 rpm for 8 sec at first and after that 3000 or 6000 rpm for 40 seconds. The thickness of polyimide films were 320 nm and 620 nm corresponding to the high rotation speeds of 6000 rpm and 3000 rpm, respectively. After the spin-coat deposition of the PI, the PI films on the n + Si substrates were subsequently cured at $100^{\circ}C$ for 10 min, 120°C for 30 min, and 200°C for 60 min on a hot-plate in the air. The thickness of the PI gate insulator was measured using ellipsometry. After these gate insulators were formed, the pentacene films for the semiconductor layers with the thickness of approximately 70 nm were deposited by thermal vacuum deposition at an estimated deposition rate of $0.2 \,\mathrm{nm/s}$ under the base pressure below 1.0×10^{-3} Pa. The pentacene was purchased from Wako Pure Chemical Industries, Ltd., and was used without further purification. Then, an Au film was thermally deposited through a shadow mask to form the source and drain electrodes. The channel length and width formed were 50 µm and 2 mm, respectively. Note that the surface of the SiO₂ prior to pentacene deposition were used without special treatment.

The transistor characteristics were measured in a vacuum chamber (FP-1500C, Japan Micronics, Inc.) using the Semiconductor Parameter Analyzer, HP4145A. The measurements of these transistors were performed in the vacuum chamber which was filled with air or evacuated at the pressure below 1.3 Pa.

RESULTS AND DISCUSSIONS

Figure 2 shows the relationships between drain–source current (I_D) and drain–source voltage (V_{DS}) at different gate–source voltages (V_{GS}) from $0\,\mathrm{V}$ to $-50\,\mathrm{V}$ for the OTFT with the $300\,\mathrm{nm}$ -thick SiO_2 gate insulator. Figure 2(a) corresponds to the I_D – V_{DS} relationships measured in the air, and Figure 2(b) corresponds to those in the vacuum pressure less than $1.3\,\mathrm{Pa}$ for the same device. As seen in the figures,

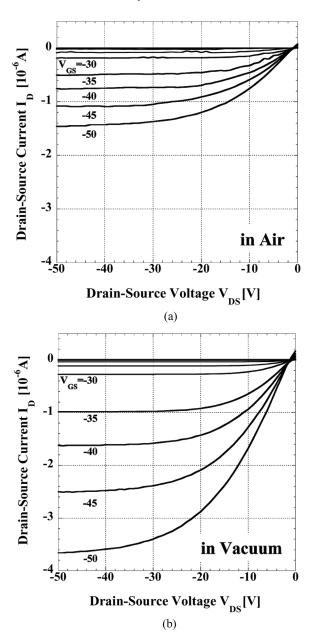


FIGURE 2 I_D – V_{DS} characteristic of Si/SiO₂ (300 nm)/pentacene/Au OTFT measured in the air (a) and in the vacuum below 1.3 Pa (b).

p-channel field-effect-transistor characteristics were obtained for the voltage ranges of V_{DS} and V_{GS} between $0\,\mathrm{V}$ to $-50\,\mathrm{V}$. As clearly seen, the drain current measured under the vacuum is approximately 2.5 times larger than that in the air. These strange behaviors were

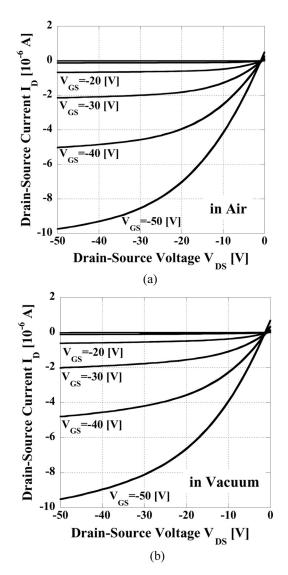


FIGURE 3 I_D – V_{DS} characteristic of Si/PI (320 nm)/pentacene/Au OTFT measured in the air (a) and in the vacuum below 1.3 Pa (b).

repeatedly reproduced with the alternate changes of the OTFT measurement environments.

Figure 3(a) corresponds to the I_D – V_{DS} relationships of the OTFT with the PI thickness of 320 nm measured in the air, and Figure 3(b) corresponds to those of the same device in the vacuum. Note that those characteristics are almost independent of the ambient pressure. Figure 4(a) corresponds to the I_D – V_{DS} relationships of the OTFT

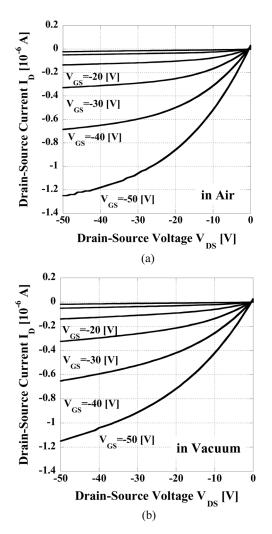


FIGURE 4 I_D – V_{DS} characteristic of Si/PI (620 nm)/pentacene/Au OTFT measured in the air (a) and in the vacuum (b).

with the PI thickness of 620 nm measured in the air, and Figure 4(b) corresponds to those of the same device measured in the vacuum. Note that those characteristics are also almost independent of the ambient pressure.

Figure 5 shows the relationships between the square root of I_D and V_{GS} for those OTFTs investigated in this work. The field-effect mobility, μ and the threshold voltage, V_{th} of each OTFT can be estimated using the following Eq. (1).

$$\mu = \frac{2L \cdot I_D}{W \cdot C_i \cdot (V_{GS} - V_{th})^2} \quad [\text{cm}^2/V \cdot s] \cdot \cdot \cdot \cdot \cdot (1)$$

where μ , L, W, C_i correspond to the hole mobility, channel length, channel width, and capacitance of the gate insulator, respectively. The derived hole mobilities, μ and threshold voltages, V_{th} are summarized in Table 1. The threshold voltage V_{th} of OTFT with the SiO₂ gate insulator decreased from approximately $-18\,\mathrm{V}$ to $-20\,\mathrm{V}$ when the measurement ambient pressure decreased, while the threshold voltages V_{th} of the OTFT with the 620 nm-thick PI films was approximately $-10\,\mathrm{V}$ independent on the PI thickness and the ambient

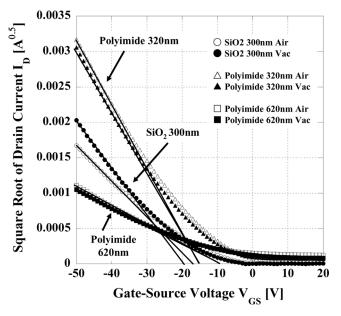


FIGURE 5 Root (I_D-V_{GS}) characteristics of pentacene OTFTs with different gate insulators measured in the air and in the vacuum.

	SiO_2 (300 nm)		Polyimide (620 nm)		Polyimide (320 nm)	
	In air	In vacuum	In air	In vacuum	In air	In vacuum
Mobility [cm ² /Vs]	9.76×10^{-3}	2.15×10^{-2}	9.76×10^{-3}	9.00×10^{-3}	5.13×10^{-2}	5.01×10^{-2}
Threshold voltage [V]	-18	-20	-10	-10	-15	-15
Drain current $[\mu A]$ $V_{GS} = -50[V]$,	-1.46	-3.66	-1.25	-1.15	-9.74	-9.52
$V_{DS} = -50[V]$						

TABLE 1 Comparison of Mobility and Threshold Voltage of the Pentacene OTFTs for Different Gate Insulators

pressures. The threshold voltage V_{th} of the OTFT with the 320 nm-thick PI films was approximately $-15\,\mathrm{V}$ independent of the PI thickness and the ambient pressures. It is noted that the mobility of the SiO_2 OTFT shows a notable decrease when measured in the air. In contrast, the mobilities of the PI OTFTs do not change with the measurement ambient pressures. The mobility of the PI OTFTs decreased with the increase of the PI thickness, and the threshold voltage V_{th} increased with the increase of the PI thickness. This may indicate the presence of negative charges in the PI gate insulator, and the charges scatter holes in the channel of the OTFTs. These effects are more significant for the OTFT with the thicker PI films.

The reduction of the threshold voltage V_{th} of the $\mathrm{SiO_2}$ OTFT when the ambient pressure decreases seems to be related to the reduction of negatively charged species from the $\mathrm{SiO_2/pentacene}$ interface. The strange behavior of the OTFT with the $\mathrm{SiO_2/pentacene}$ interface on the ambient pressure is unknown. This might be related to the facts that the surface of $\mathrm{SiO_2}$ is hydrophilic while the PI surface is hydrophobic. It was also speculated that the amounts of absorbing and desorbing of some chemical species from the interface of the pentacene/polyimide could be much smaller compared to those from the interface of the pentacene/ $\mathrm{SiO_2}$. Further investigations are needed to conclude the mechanisms of these behaviors.

CONCLUSION

The *p*-channel pentacene organic thin film transistors (OTFTs) with two different kinds of the gate insulators of the SiO₂ and the polyimide were fabricated, and the transistor characteristics were measured in the air and in the vacuum below 1.3 Pa and compared. The hole

mobility of pentacene OTFT with the SiO₂ gate insulator measured under the vacuum is approximately 2.5 times larger than that measured in the air. However, the hole mobility of the pentacene OTFTs with the polyimide gate insulators measured is almost independent of the measurement environmental pressures. It was speculated that the amounts of absorbing and desorbing of some chemical species from the interface of the pentacene/polyimide could be much smaller compared to those from the interface of the pentacene/SiO₂.

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